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Coupling Heat and Electricity Sources to Intermediate Temperature Steam Electrolysis

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Abstract: The use of CO_2-free energy sources for running SOEC (solid-oxide electrolysis cell) technologies has a great potential to reduce the carbon dioxide emissions compared to fossil fuel based technologies for hydrogen production. The operation of the electrolysis cell at higher temperature offers the benefit of increasing the efficiency of the process. The range of the operating temperature of the SOEC is typically between 800 °C and 1,000 °C. Main sources of degradation that affect the SOEC stack lifetime is related to the high operating temperature. To increase the electrolyser durability, one possible solution is to decrease the operating temperature down to 650 °C, which represents the typical operating range of the ITSE (intermediate temperature steam electrolysis). This paper is related to the work of the JU-FCH project ADEL, which investigates different carbon-free energy sources with respect to potential coupling schemes to ITSE. A predominant focus of the analysis is put on solar concentrating energy systems (solar tower) and nuclear energy as energy sources to provide the required electricity and heat for the ITSE. This study will present an overview of the main considerations, the boundary conditions and the results concerning the development of coupling schemes of the energy conversion technologies to the electrolyser.

Key words: Intermediate temperature electrolysis, electrolyser, hydrogen, solar, flow chart.

1. Introduction

Hydrogen is mainly produced from fossil fuels through steam reforming of natural gas. Unfortunately, the fossil fuel based hydrogen generation does not contribute to the reduction of greenhouse gas emissions. Hydrogen can be generated instead by carbon-free energy sources via water electrolysis in order to avoid the emissions of carbon dioxide. The concept of integrating renewable energy with hydrogen systems was given serious consideration in the 1970s [1, 2]. Numerous studies have been reported on the hydrogen production systems from solar energy, wind energy and biomass in Refs. [3-5].

There are three main types of water electrolysis: the alkaline electrolysis, the PEM (polymer electrolyte membrane) electrolysis and the SOEC (solid oxide electrolysis cell) [6, 7]. Most of the water electrolysis technologies to date have used alkaline or acidic electrolyte systems for hydrogen generation [8, 9], which are operated at temperatures below 100 °C, while the SOEC operates with water steam at temperatures in the range of 800-1,000 °C. SOEC uses

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a combination of thermal energy and electricity and the water is fed to the electrolyser in vapor phase. The advantage of the operation at higher temperature consists of reducing the electricity demand of the electrolyser. In addition to that, the reaction kinetics is increasing at higher temperatures. The overall thermal-to-hydrogen efficiency for the high temperature electrolysis can be as high as 50%, which is about double the overall efficiency of the alkaline electrolysis [10]. This higher operating temperature of the SOEC has an impact on the solid oxide cells and stack lifetime. Indeed, this temperature range of 800 °C to 1,000 °C enhances chemical species evaporation and diffusion, resulting in the formation of secondary isolating phases, as well as in a decrease of the mechanical stability of ceramic and metal components.

To increase the electrolyser durability, a very suitable solution is to decrease the operating temperature of the electrolyser. The resistances of cells, interconnects and contact layers will tend to increase and to limit the electrolyser performance, but at the same time, all parasitic phenomena such as interdiffusion, corrosion or vaporization responsible for cell and stack degradation will be significantly slowed down. Cells and interconnect coatings are now available that allow reaching at 600-650 °C equivalent performance that are obtained at 800 °C in the frame of an EU founded project [11] . The operation of the electrolyser in temperature range of 600 °C to 650 °C is known as the ITSE (intermediate temperature steam electrolysis). The reduction of the operating temperature reduction will open the door for extended operation modes and will allow the coupling of the electrolyser with renewable energy sources. As a consequence, heat sources such as solar energy or nuclear high temperature reactor could directly feed the electrolyser.

Within the ADEL project, the integration of the ITSE system with different carbon-free energy sources has been analyzed. Some of these energy sources are: CSP (concentrated solar power) systems (tower systems, parabolic trough collectors, dish Stirling and linear Fresnel reflector), nuclear plants etc.. In this work, the integration with central receiver systems using air, steam and molten salt as well as nuclear PWR (pressurized water reactor) are analyzed.

2. Thermodynamics of the Intermediate Temperature Electrolysis

2.1 Principle

ITSE aims to operate at temperatures below 700 °C. This process integrates a ceramic electrolyte conducting ions O^{2-}. The advantage of this process is that its electricity consumption is lower than the one of a low temperature electrolyser (PEM or alkaline), as seen in the thermodynamic diagram (Fig. 1). On top of that, the improvement of reaction kinetics at high temperature enhances the process efficiency. This type of process could enable a large scale hydrogen production in the future [12].

The reactions that take place in the electrolyser are:

Cathode: $H_2O_{(g)} + 2e^- \rightarrow O^{2-} + H_{2(g)}$

Anode: $O^{2-} \rightarrow \frac{1}{2}O_{2(g)} + 2e^-$

At the cathode (hydrogen electrode), the water molecule is dissociated because of the electrical current and the heat provision, in order to form a molecule of hydrogen and an oxygen ion O^{2-}. Under the effect of the electrical field, the ion O^{2-} goes through the electrolyte and migrates to the anode (oxygen electrode) where its oxidation produces an oxygen molecule O_2.

2.2 Operation Mode

Thermoneutral mode:

The thermoneutral voltage ($E_{thermoneutral}$) of an electrolyser corresponds to an operation where electrolysis-needed heat is brought by all the overvoltages present in the cell. This voltage is given by the following equation:

$$E_{thermoneutral} = -\frac{\Delta_r H}{nF} \quad (1)$$

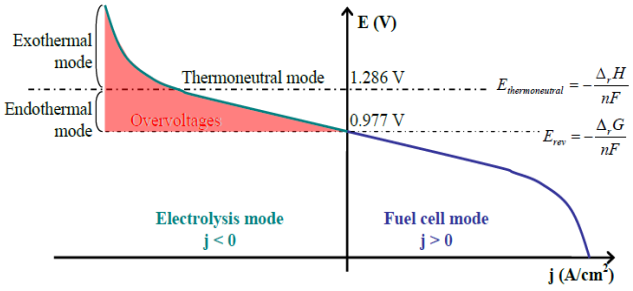

Fig. 1 Scheme of different HTSE operating modes at 1,073 K.

$E_{thermoneutral}$: thermoneutral voltage in V;

$\Delta_r H$: enthalpy change of reaction in J/mol;

F: faraday constant in C/mol;

n: number of exchanged electrons in the water electrolysis reaction.

At 298 K and 1 bar, the thermoneutral voltage is 1.481 V. It decreases to 1.286 V at 1,073 K and 1 bar. At this particular voltage, the heat absorbed by the endothermic electrolysis reaction is compensated by overvoltages. In this mode, outlet gases (H_2/H_2O and O_2) have the same temperature as the inlet gases (H_2O/H_2).

Exothermal mode:

In this operating mode, the cell voltage is above the thermoneutral voltage. Current densities imposed on the cell are higher, so that overvoltages are also higher. Heat production in the cell is higher than heat absorbed by the endothermic electrolysis reaction. Thus, exhaust gases (H_2/H_2O and O_2) have higher temperature than inlet gases (H_2O/H_2). Therefore, low temperature steam can be heated up to the electrolyser operational temperature just by the exhaust gases. As a part of the electricity is used to overheat the streams, the voltage efficiency of this mode is lower than 100%.

Enodthermic mode:

If the cells' voltage is lower than the thermoneutral voltage, overvoltages are also lower. Heat production in the cell is lower than heat absorbed by the endothermic electrolysis reaction. Therefore, outlet streams (H_2/H_2O and O_2) have lower temperature than the inlet gases (H_2O/H_2). Heat to reach the operational stack temperature should be provided by external heat.

As a part of the heat needed by the endothermic reaction is supplied by external heat sources, the voltage efficiency of this mode is higher than 100%. This operating mode is particularly adapted for coupling a high temperature electrolyser with an available external high temperature heat source.

2.3 System Performance

Theses electrolysers can operate at high current densities (up to 1.4 A/cm^2 with 1.1 V at 800 °C [13] and up to 3.6 A/cm^2 with 1.48 V at 950 °C [14]). Because they are at research state, most tests have been carried out only at atmospheric pressure on planar cells so far. Tubular cells operate better at higher pressure, but research is currently focuses on planar cells, whose production capacity can be higher. The operating temperature is commonly 600-800 °C. The system can reach 70% efficiency based on primary energy (considering that the production of electricity has an average efficiency of less than 40%), or 90% efficiency based on total energy input into the electrolyser [15]. The ITSE operating temperature enables ionic conductivity of the electrolyte to be high enough, and ensures high electrode reaction kinetics, so that electrode overvoltages are low compared to low temperature electrolysis.

These different elements have a notable impact on the total electricity consumption of electrolysis. Indeed, the electricity consumption is around 4.6 kWhe/Nm^3 of hydrogen with low temperature electrolysis, whereas it is 3.2 kWhe/Nm^3 of hydrogen with high temperature electrolysis in the thermoneutral mode. The

consumption can even be reduced to 2.6 kWhe/Nm^3 of hydrogen in the endothermic mode according to the results of Dönitz et al. [16].

3. Energy Conversion Technologies

3.1 Solar Tower Applying Air as Heat Transfer Fluid

3.1.1 Description of the Technology

Some current European designs use air as heat transfer medium for solar power tower operation because of its high temperature and its good handiness [17]. Another advantage of air as heat transfer medium is its abundance without costs, his non-toxicity, and the fact that it does not require freeze protection during times of non-operation. That is why air is also considered as heat transfer fluid in the present concept for powering an ITSE system. Ambient air is sucked through a blackened porous ceramic structure on which the solar radiation is focused. The air cools the outer parts of the receiver and is heated up gradually to the design temperature level at the inner surface of the porous absorber. In passing through the receiver, the air is heated up to 700 °C and this heat is delivered to the water-steam cycle in a heat recovery boiler. The steam generated there drives a turbine/generator and returns in form of condensate to the steam generator (Fig. 2).

3.1.2 Process Design Basis

In this study, a solar tower with a nominal capacity of 10 MWe is considered. The time-dependant performance for the solar collector field and the solar transients and fluctuations can be resolved by using an oversized heliostat field and making use of the excess energy to load a thermal storage system. The electricity for the electrolyser will be generated with a steam turbine for which the steam will be produced with concentrating solar energy. Power tower plants use a conventional power block and can easily dispatch power when storage is available. The thermal source of the steam generator is hot air stream at 680 ºC.

Thermal-energy storage in the power tower allows electricity to be dispatched to the grid when demand for power is the highest, thus increasing the monetary

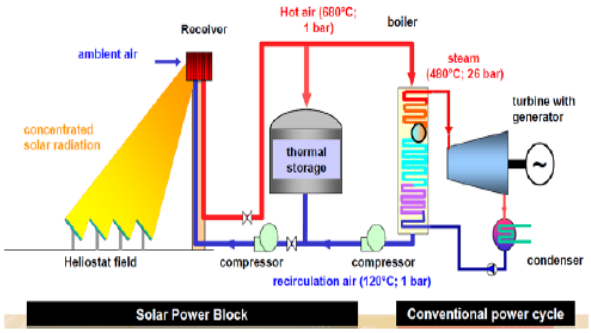

Fig. 2 Air solar power tower.

value of the electricity. Besides making the power dispatchable, thermal storage also gives the power-plant designer freedom to develop power plants with a wide range of capacity factors to meet the needs of the utility grid.

In parallel to the steam generator and receiver, the heat storage is integrated into the power cycle and implemented as air-cooled regenerator storage. This technology stores in a ceramic solid-state storage system some of the thermal energy collected by the solar field for conversion to electricity later in the day. Storage can adapt the profile of power produced throughout the day to demand and can increase the total power output of a plant with given maximum turbine capacity. With this storage type, air is in direct contact with the solid storage medium and exchanges heat as it flows along a flow-path through the storage medium.

3.1.3 Flow Chart

The solar power tower consists of the following components:

- an heliostat field;
- an open volumetric receiver (RECEIVER), at the top of the solar tower, for air heating and sweep gas generation.

The electrolyser consists of two components:

- a steam splitter (STEAMSPL), where the water splitting reaction will take place;
- a membrane (MEM), where the product is splitted into a stream containing H_2 and H_2O and a stream containing O_2 and N_2.

Finally, the heat recovery system is used to overheat the water steam up to the operating temperature of the

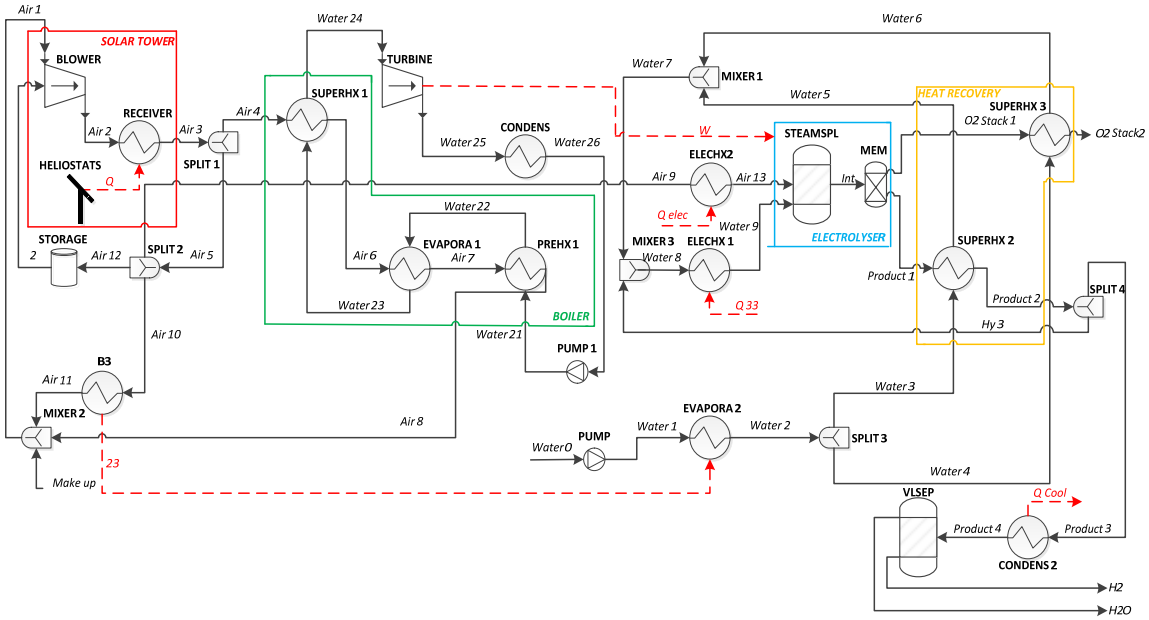

Fig. 3 Flow chart of the air cooled solar tower coupled to the ITSE.

electrolyser. It consists of two heat exchangers (SUPERHX 3, SUPERHX 2).

The above figure (Fig. 3) shows the flow chart of the plant.

High concentrated solar radiation is used in the open volumetric receiver (RECEIVER) to produce hot air at 700 °C (Air 3) to generate high temperature steam in a heat boiler (BOILER). This steam is used as feed for a steam Rankine cycle (TURBINE) to generate electricity for the electrolyser.

A part of the hot air is directly delivered to the electrolyser and used as sweep gas (Air 9) while another part (Air 12) is led to the storage system (STORAGE) which is connected parallel to the boiler. A third part of the hot air production (Air 10) is used for the evaporation of the electrolysis water. The evaporator has been simulated as a single heat exchanger (EVAPORA 2), which is connected to B3 by the red heat stream (23) in the flow diagram.

After the evaporation of the electrolysis water, the steam (Water 2) is split in the splitter (SPLIT 3) into two sub-streams (Water 3 and Water 4); the first sub-stream (Water 3) is overheated in the heat exchanger (SUPERHX 2) up to 680 °C by the stream (HY1) leaving the electrolyser. The second sub-stream (Water 4) is overheated in the heat exchanger (SUPERHX 3) up to 525 °C by the stream (O2 Stack 1). Then, both sub-streams are mixed in the mixer (MIXER 1) and the mixture is introduced to another mixer (MIXER 3), where it will be mixed with the stream (HY 3) in order to maintain reducing conditions at the cathode side, since it was assumed by the ADEL project-partner that the stream (Water 8) has to contain 10 mol% H_2.

An electrical heater (ELECHX) is necessary in order to rich the operating temperature of the electrolyser. A second electrical heater is added to heat the sweep gas up to 700 °C if it is necessary for the transient conditions. Thus it enables to control the sweep gas temperature and to guarantee fixed conditions for the sweep gas in the electrolyser. The electrical air and steam heaters are modelled as heaters receiving heat, which is considered to be a realistic representation since virtually all electric power supplied will be dissipated as heat.

The first simulation results show that the quantity of sweep gas and the ratio sweep gas/steam is a very important factor for the process efficiency. The 10

MWe plant with a sweep gas flow rate of 16.6 kg/s can produce 3,240 Nm^3/h hydrogen. In an optimized case, an efficiency of 62% has been reached.

3.2 Direct Steam Generation Central Receiver System, DSG-CRS

Among central receiver concentrating solar plant, water/steam cooled receiver technology is the most mature, with commercial plants operating since 2007. DSG-CRS has the advantage of avoiding the steam generator required in other CSP technologies, which might reduce the cost of the plant. Within this study the possibility of integration of an ITSE into a DSG-CRS has been analyzed. The variables analyzed in the study were the electrolyser steam generation system, thermal energy storage, and steam conditions and design of the Rankine cycle.

As can be seen in Fig. 4, the feed water stream required by the ITSE can be preheated and superheated up to 660 ºC by the heat recovery system of the exhaust gases. Thus, external heat input is limited to the evaporation step, and a last superheated step to increase the stream inlet temperature to the nominal operational temperature of the electrolyser. On the other hand, the sweep gas stream, air in this case, is heated by the exhaust sweep stream up to 680 ºC, requiring a last heater to overcome the driven temperature difference of the heat recovery system. Both streams include an electrical heater to carry out the last heating step. To evaporate the water stream two options were proposed, direct evaporation into the receiver at the top of the tower, and using steam extracted from the power block as heat source of a heat exchanger. A low fraction of steam is extracted from the low pressure turbine, and sent to a thermal PCM (phase change material) storage tank/steam generator. Steam is there condensed, and the PCM melted. Through a secondary line, the electrolyser water stream is fed at 95 ºC, and evaporated by the solidification of the PCM. Through this system a higher temperature difference between steam and water is required, but heat is stored at the same time. Thereby the hydrogen production plant is able to work even when the DSG-CRS plant is shut down, running with power from the grid. Thus, the power/hydrogen production plant can be used as an active grid buffer.

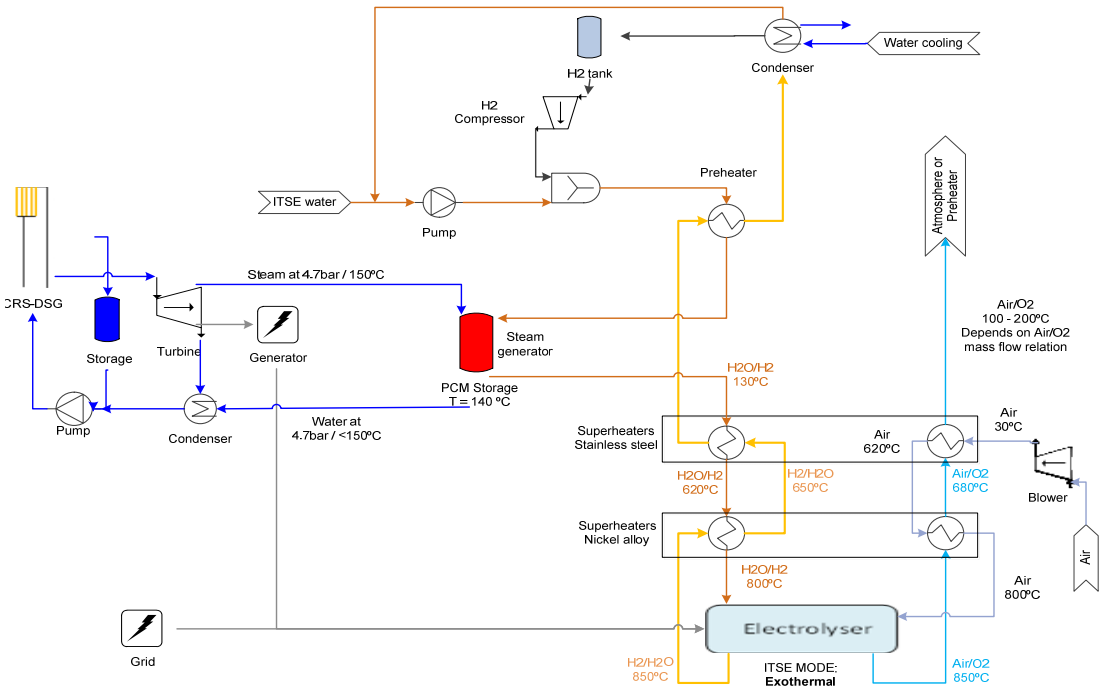

Fig. 4 Flow chart of the integration of the ITSE into a DSG-CRS.

Seven different Rankine cycles have been compared for DSG-CRS plants which capacities are 10 MWe and 50 MWe. Steam conditions have been varied from saturated to superheated condition; and reheat and no-reheat turbine have been analyzed. In every case, the reduction of the DSG-CRS performance when integrating the ITSE unit has been lower than 4%.

Under nominal conditions, the electrolyser unit achieves an electrical efficiency of 94.7%. Taken into account thermal requirements for the feeding water evaporation, the efficiency of the unit decreases down to 75.3%. The ITSE unit required about 35 kWh of electricity and 9 kWh of heat to produce 1 kg/h of hydrogen. The small CSP plant, 12.4 MWe, coupled with two ITSE units of 2.5 MWe, is able to produce 1,656 Nm^3/h of hydrogen, and inject to the grid above 7 MWe. The bigger plant, with a total electricity production of 62 MWe, was coupled with three ITSE units of 10 MWe, producing 9,938.5 Nm^3/h of hydrogen and injecting to the grid more than 31 MWe.

From the simulation it was found that the Rankine cycle performance variation decreases by just 2.6% when the ITSE Unit is integrated; and the hybrid plant has 5.5% lower performance in comparison with the reference solar plant without the electrolyser.

3.3 Molten Salt Tower System

The third option analyzed was the integration of the ITSE unit with a molten salt tower plant. Molten salt-cooled central receiver plant has two heat transfer fluid loops, which decouples the steam generation from the collector subsystem and makes possible the integration of high capacity thermal energy storage. Therefore, the ITSE can operate under high stable steam and electricity conditions. Thanks to long-term thermal storage system, which normally is integrated in these kinds of plants, it is possible to operate round-the-clock in summertime, leading to an annual capacity factor of 74%. Therefore, these plants can be designed easily for both cases: stand-alone hydrogen production plants or grid stabilizer as a power sink at low demand periods.

The temperature range of this kind of plants is between 565 ºC in the hot tank, and 290 ºC in the cold tank. The cold tank is kept 70-40 ºC above the salt mixture freezing point, 220-250 ºC. Steam temperature at the inlet of the electrolyser system must be at 130 ºC. Therefore, it could be possible to feed the ITSE steam generator with cold molten salt. In this way, temperature difference between hot and cold streams of the steam generator is reduced, maximizing the energy efficiency of the process. However, it should be kept in mind that solidification must be avoided and therefore, molten salt can not be cooled down below a limited temperature, higher than freezing temperature. The temperature reduction achieved by the cold molten salt through the steam generator depends on the salt mixture quality, its design cold temperature and the required steam mass flow. The proposed scheme for the integration of the ITSE into a molten salt power plant is shown below on the Fig. 5.

A full hydrogen dedicated molten salt tower plant of 30 MWe and a solar multiple of 1.3 have been analyzed at nominal conditions. For the hybridization, the molten salt from the cold tank is firstly sent to the electrolyser evaporator, and then to the solar receiver. The temperature of the cold tank has been increased from 290 ºC to 300 ºC to be able to keep the temperature of the salt after the electrolyser evaporator at 270 ºC. As result of this variation, the mass flow of molten salt that can be heated up within the solar receiver is reduced by 4.3%. Concerning the electrolysis process, it consists of a 29 MWe stack, with 1 MWe of parasitic loads, mainly in the superheaters. The hybrid plant is able to produce 9,575 Nm^3/h of hydrogen.

3.4 Nuclear PWR (Pressurized Water Reactor)

Another energy source selected consists of a nuclear reactor, which has a high degree of availability and is a non-intermittent source of energy. Coupling a reactor and an electrolyser requires installing a power block

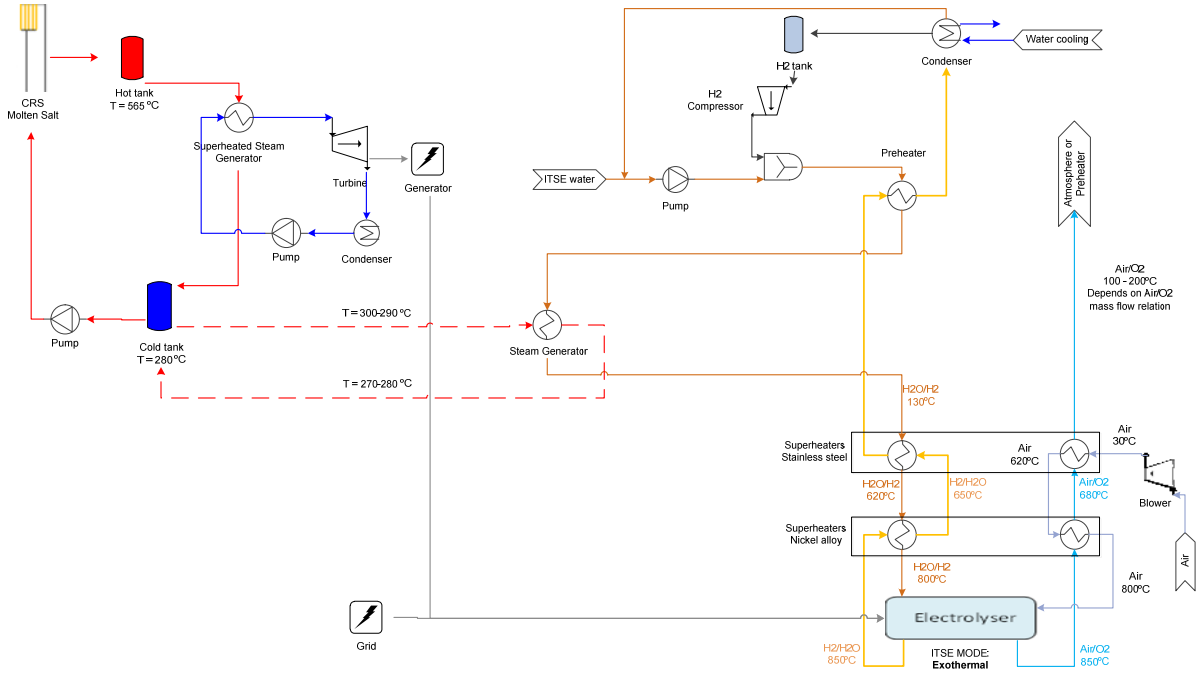

Fig. 5 Flow chart of the integration of the ITSE into a molten salt solar tower.

that uses part of the heat from the reactor to generate the electric power needed for the electrolyser. Therefore, a proper power block needs to be chosen, and this decision is mainly determined by the reactor technology chosen and by the way of coupling it with the electrolyser system.

A PWR was chosen, since it is a mature technology being able to reliably provide the necessary electricity and heat to generate the steam. A typical steam cycle is considered for a PWR reactor that will extract the electricity demanded by the electrolyser. Furthermore, in this case, the only feasible option is to transfer heat to the electrolyser system from one of the extractions of the low pressure turbine in that cycle, and choosing a suitable pressure for it accordingly. It will therefore be necessary to have an electrolyser system that includes heat recovery and electric heating in order to reach the 700 ºC as required.

It would be a steam cycle operating with saturated steam at 70 bar at the inlet to the high pressure turbine, where it would expand up to 12 bar. This would also be the pressure of the deaerator fed from the outlet of that HP turbine. In addition, the discharged steam would go to the required moisture separator (the steam would exit the high pressure turbine at too low a quality to keep it expanding in the low pressure turbine), and then undergo double heating with steam taken from the high pressure turbine and with main steam. The reheated steam then goes to the low pressure turbine and condenses. A pump pumps the condensate through the train of low pressure pre-heaters and on to the deaerator. The deaerator drainage is pumped by the feedwater pump to the train of feedwater pre-heaters and finally enters the reactor steam generator. To do this, a configuration was chosen consisting of two extractions from the high pressure turbine (two high pressure pre-heats) and three extractions from the low pressure turbine to do three low pressure pre-heats. In addition, there is also a first extraction just at the inlet to the turbine for the thermal demand of the electrolyser system. The most efficient way to produce H_2 using a constant power source, as a nuclear reactor, is to feed.

Connecting the power cycle to the grid, which will absorb a potential surplus of energy, allows to couple hydrogen production from nuclear energy as a constant power source to a variable sink of energy like storage.

At the given electrolyser operation temperature (700 ºC), the ratio of heat to electricity required is 20 to 80. The molar ratio of hydrogen in the product stream is 63% at the electrolyser outlet, which means that 7,823.4 Nm^3/h of hydrogen are produced by the chemical plant.

The proposed scheme for the integration of the ITSE into a nuclear pressurized water reactor is shown below in Fig. 6.

4. Conclusions

The coupling of solar central receiver systems and the nuclear pressurized water reactor with an ITSE unit has been analyzed. The solar tower technology with ambient air as heat transfer fluid has been analyzed. As the air is heated in the receiver up to similar temperature than the ITSE operational temperature, 700 ºC, it can be used directly as sweep gas in the electrolyser. Central receiver systems with direct steam generation have been investigated. The integration of the ITSE into the solar power plant has a low influence, reducing less than 4% the performance of the DSG-CRS. Regarding molten salt tower plants, the results show that, if the reference temperature of the cold tank is set at 300 ºC, it is possible to use the cold salt to evaporate the electrolyser feeding water without reducing the temperature of the salt below 270 ºC. In this case, that the solar receiver inlet temperature of the molten salt is lower, the mass flow of the molten salt that is heated up within the receiver is decreased by 4%. With respect to the nuclear energy source, a conventional light water reactor (PWR) has been studied. It is concluded that the most effective way to produce hydrogen using a constant power source, as a nuclear reactor, is providing energy to the grid and the hydrogen plant at the same time. With this design, it would be possible, if required, to regulate the output to the grid by varying the production of hydrogen. In the following phase of work, it will be necessary to analyze the influence of transients on process performance and on operational strategies by dynamic simulation. To be able to analyze the prospects of the technology most concretely, it will be meaningful to select representative scenarios with a suitable site of this application and its boundaries conditions. For such cases, the set-up of the real plant and the evaluation of

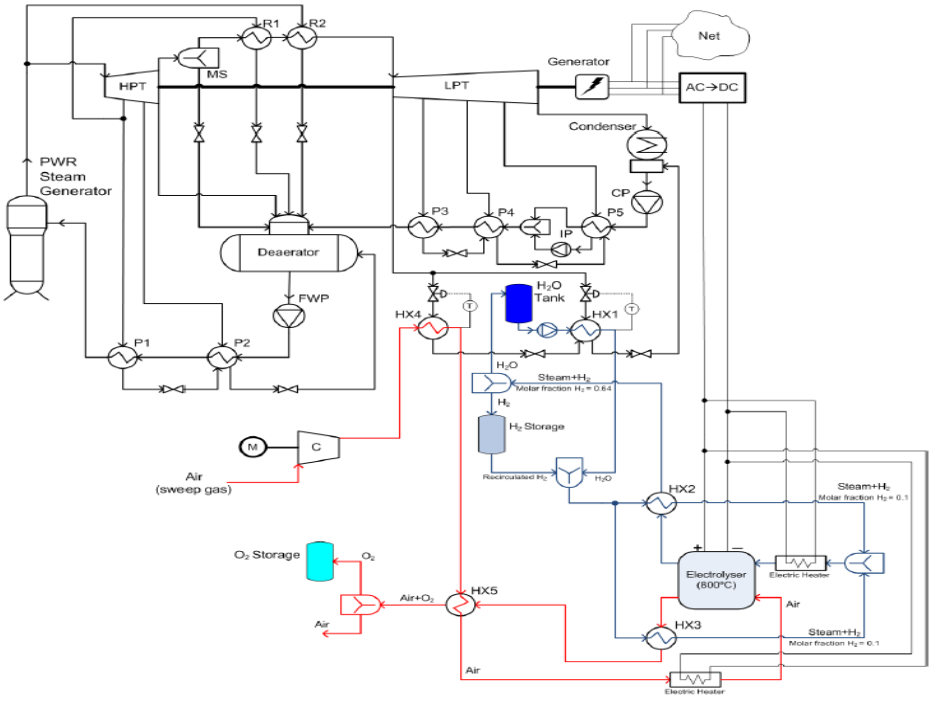

Fig. 6 Flow chart of the integration of the ITSE into a PWR reactor coupled to the steam cycle.

energy efficiencies and performance under transient conditions shall be analyzed to get a refined view on the future potential of the technology.

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